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Eddy covariance measurements of sea spray particles over the Atlantic Ocean

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Most estimates of sea spray aerosol source functions have used indirect means to infer the rate of production as a function of wind speed. Only recently has the technology become available to make high frequency measurements of aerosol concentration suitable for direct eddy correlation determination of the particle flux. This was accomplished in this study by combining a newly developed fast aerosol particle counter with an ultrasonic anemometer which allowed for eddy covariance measurements of size-segregated particle fluxes. The aerosol instrument is the Compact Lightweight Aerosol Spectrometer Probe (CLASP) – capable of measuring 8-channel size spectra for mean radii between 0.15 and 0.35 μm at 10 Hz. The first successful measurements were made during the WASFAB (Waves, Air Sea Fluxes, Aerosol and Bubbles) field campaign in October 2005 in Duck (NC, USA). The method and results are presented and comparisons are made with recent sea spray source functions from the literature.

1 Introduction

Sea spray particles are salt water droplets ejected from the ocean. The aerosols formed from sea spray particles are important because they have a significant impact on climate processes, both directly via the scattering of solar radiation, and indirectly via their influence on cloud microphysical properties. Sea salt aerosol are the single most important factor controlling the scattering of radiation, and hence the radiation budget near the surface, over the open oceans (Haywood et al., 1999); they dominate the particulate mass concentration in unpolluted marine air, and contribute approximately 44% to the global aerosol mass fluxes (Seinfeld and Pandis, 1998). They act as cloud condensation nuclei (CCN) and have a large influence on both the microphysics and chemistry of marine stratocumulus clouds (O’Dowd et al., 1999), which are one of the largest sources of uncertainty in climate predictions. They have a large size range, with radii from 0.01 μm up to 1 mm radius (Martenssen et al., 2003; Clarke et al., 2006).

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Sea spray aerosol droplets are produced by different mechanisms. Jet and film droplets are produced from bursting bubbles. Bubbles form predominantly from breaking waves, but there are many other possible mechanisms for producing bubbles: biological processes, de-gassing of air-rich surface layers as the water is warmed, volcanic release of gases, and rain falling on the ocean surface. For 10-m wind speeds above about 5 m s^{-1} the stress of the wind on the ocean surface is sufficient to cause the water on the surface to move faster than that underlying it, forming a wave that breaks under gravity (Lewis and Schwartz, 2004). As they break, the waves entrain air into the ocean surface producing a plume of bubbles (Blanchard and Woodcock, 1980). As the bubbles reach the ocean surface, patches of white foam form – whitecaps. Whitecaps have been classified into two stages. As a wave breaks, it is capped at the sea surface by a bright stage A whitecap (Monahan, 2002); this has a small surface area and are rich in bubbles. Stage A whitecaps have a microwave emissivity of approximately 1 (Monahan, 2002) compared to an emissivity of 0.5 for unperturbed ocean surfaces. As the bubbles in the whitecap burst, the decaying foam patch becomes much less distinctive, appearing grey rather than white. This is a stage B whitecap; it contains fewer bubbles than stage A whitecaps, and has an emissivity intermediate between that of unperturbed water and a stage A whitecap, decreasing as the bubbles burst. Stage B whitecaps are less easily identifiable by remote sensing instruments and photographic surveys, introducing a source of uncertainty into attempts to quantify the impact of whitecap fractional area on various air-sea exchange processes (Anguelova and Webster, 2006).

As a bubble starts to protrude above the ocean surface, some of the liquid film surrounding the bubble drains off, thus thinning and weakening the film. The film ruptures and rolls back on itself before producing film droplets (Blanchard and Woodcock, 1980; Spiel, 1998). Jet droplets are produced when the pressure is released from inside the bubble as it collapses. A liquid jet shoots up from the base of the bubble cavity, due to hydrostatic forces. The jet then quickly becomes unstable and breaks into a number of jet droplets (Blanchard, 1983). Spume drops are produced in situations where the wind

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stress is of sufficient magnitude to physically rip droplets of water from the wave crests, ejecting them horizontally into the air stream over the wave. Typically, this occurs for wind speeds exceeding 9 m s^{-1} (Monahan et al., 1983). Spume drops are the largest of the sea spray droplets, measuring up to 1 mm in radius (Fairall et al., 2006).

5 The sea spray source function (SSSF) describes the amount (number, volume, mass) of sea spray aerosol produced at the sea surface per unit of time and per surface area as a function of environmental conditions. The need for an accurate SSSF over a wide range of environmental conditions has been emphasised by global climate studies (IPCC, 2001) and the fact that sea spray particles are the second largest single source
10 of aerosol mass injected into the atmosphere (Seinfeld and Pandis, 1998). They are especially important as a source of cloud condensation nuclei (CCN), thereby affecting the chemistry and radiative effects of marine clouds.

SSSF's available in the literature vary by as much as 6 orders of magnitude (Andreas, 1998, 2002). However results from recent work in the sub-micron area converge
15 to within about a factor of 2 (Clarke et al., 2006). Many of the methods used to derive the source functions are indirect, relying either on an assumption of steady state in the atmosphere, under which the production flux equals a known deposition flux (e.g., Andreas, 1992; Smith et al., 1993; Fairall et al., 1994; Smith and Harrison, 1998), or using the whitecap method which combines parameterizations of the fractional white-
20 cap cover based on field observations with laboratory measurements of the sea spray aerosol production rate per unit whitecap (e.g. Monahan et al., 1986; Mårtensson et al., 2003). For recent formulations of the sea spray source function, see Schulz et al. (2004) and O'Dowd and De Leeuw (2007).

25 There are potential problems associated with the assumptions of steady state leading to balance between production and removal of sea spray particles (e.g., Fairall et al., 1994; Smith et al., 1993; Smith and Harrison, 1998). A direct measurement of the aerosol flux would eliminate most of these problems, and would be expected to result in a physically more robust source function. The ideal approach to any flux turbulent measurement is eddy covariance. This method has been used to measure particle

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deposition to forests (Gallagher et al., 1997; Buzorius et al., 1998) and recently also to look at the production flux of sea salt particles (Nilsson et al., 2001; De Leeuw et al., 2003; Geever et al., 2005). The eddy covariance method correlates turbulent fluctuations of the vertical wind component with those of particle concentration. The vertical wind speed is usually measured with an ultrasonic anemometer. The vertical turbulent flux is obtained from the equation for total vertical flux via Reynolds averaging (see Stull, 1988). The sign of the flux indicates an upward (positive) or downward (negative) transport of the quantity of interest. In flux calculations, it is usual to transform the measured wind components into a coordinate system such that \bar{u} , the component along the x-axis, is aligned with the mean streamline, and \bar{v} and \bar{w} – the mean horizontal and vertical cross-streamline components respectively – are both equal to zero. Application of the EC method to measure the production of sea spray particles requires an instrument that can measure aerosol concentrations with a high temporal resolution – eddy correlation requires a minimum sample rate of 2–3 Hz – combined with a high sample volume in order to achieve robust statistics with low ambient particle concentrations. Such direct measurements of sea spray fluxes have been possible very recently, as instruments with sufficiently high sample rates have become available, but remain sparse due to the bulky and expensive nature of most of the aerosol instrumentation, which makes it awkward to use close to the sea surface without either causing flow distortion or risking damage to the instrumentation in a hostile environment. The eddy covariance method was first applied to the measurement of sea spray fluxes by Nilsson et al. (2001), who assembled a flux package consisting of a sonic anemometer and a Condensation Particle Counter (CPC). The CPC used had a temporal resolution of 3 Hz and measured the total particle concentration for radii larger than 10 nm. This flux package was expanded by De Leeuw et al. (2003), with an optical particle counter with a heated inlet to allow for size segregated measurements and the selective sampling of sea salt particles via the volatility technique by heating the sample to 300°C (De Leeuw et al., 2003). Geever et al. (2005) used this flux package to determine fluxes in two size ranges.

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The objective of this paper is to demonstrate the feasibility of size-resolved sea spray flux measurements using the eddy covariance method with a novel instrument developed at the University of Leeds couples a high sample volume to fast time response. The Compact Light-weight Aerosol Spectrometer Probe (CLASP) version used here can measure particles with radii in the range $0.15\text{ }\mu\text{m}$ to $3.5\text{ }\mu\text{m}$. It has a flow rate of $50\text{ cm}^{-3}\text{ s}^{-1}$ and a sample rate of 10 Hz. The main component of CLASP is a MetOne Optical Particle Counter (OPC) from Pacific Scientific Instruments. Particles in the MetOne sample volume scatter light from a laser (wavelength 780 nm). The light scattered in the side lobes is detected by a photodiode; its intensity is a measure of the particle's size. In CLASP the MetOne OPC is connected to an 8 channel pulse height analyser to classify the peaks in output signal into 8 size ranges. See Hill et al. (2007)¹ for more details about CLASP.

The CLASP instrument is especially suitable for in-situ flux measurements in combination with an ultrasonic anemometer because of its small size (approximately $11\times 15\times 6\text{ cm}$) and light weight, which allows co-location with the sonic anemometer without causing significant flow distortion, and thus minimizing the length of inlet tube (here approximately 0.5 m) and hence particle losses. A high flow rate ensures that adequate statistics can be achieved to characterize spectra within a time period of approximately 10 min, similar to the averaging time required for surface layer flux measurements.

2 Field campaign

The Waves, Air Sea Fluxes, Aerosol and Bubbles (WASFAB) experiment (De Leeuw et al., 2007; Zappa et al., 2006) took place during October 2005 at the Army Corps of Engineers Field Research Facility (FRF) in Duck, North Carolina. The aim of the overall

¹Hill, M., Brooks, B., Norris, S. J., Smith, M. H., De Leeuw, G., and Lingard, J.: A novel high-temporal resolution particle spectrometer, J. Atmos. Ocean Technol., in review, 2007.

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project was to obtain a large set of measurements to help constrain the sea spray source function. The location in North Carolina was selected because of the 560m long pier and supporting oceanographic and meteorological observations available at the site. The measurement site was located at the end of the pier, well beyond the surf zone. Initial micrometeorological measurements made at the same location during the winter of 2005/2006 demonstrated that the local drag coefficient compared favourably with that obtained from the TOGA-COARE bulk flux parameterization (Zappa et al. 2006); we thus have confidence that the site is representative of the open ocean under onshore wind conditions (directions from 5° to 120°).

The flux package used here (Fig. 1) consisted of a Gill R3A ultrasonic anemometer and CLASP, and was situated on a lattice tower at 16.5 m above the mean sea surface at the end of the pier. The measurements were supported by a second aerosol flux package consisting of an ultrasonic anemometer, CPC and OPC with a heated inlet at 300°C and a Licor LI-7500 open path sampler for water vapor and CO₂ fluxes. Supporting instruments included a sea spray package consisting of a PMS FSSP, OAP and PCASP, along with an aethalometer to provide information on the absorbing aerosol in the air mass. Meteorological stations provided information on the local temperature, humidity, wind speed and wind direction; offshore buoys provided wave measurements.

Flux losses due to displacement between the sonic anemometer and the CLASP inlet were calculated using the method described in Kristensen et al. (1997) to be 0.2%. The time delay between a particle entering the CLASP inlet tube and entering the MetOne sample volume was calculated to be 0.2 seconds; this is in good agreement with the 2-sample offset obtained by determining the sample offset required to maximize the covariance between the turbulent vertical velocity and the CLASP concentrations. This offset was applied to the CLASP time series prior to calculating the turbulent fluxes.

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3 Data processing

When taking spot measurements as a function of time in a turbulent field, Taylor's hypothesis is often used. This states that eddies change imperceptibly as they are convected by the mean wind past an in-situ sensor (Kaimal and Finnegan, 1994). Under Taylor's hypothesis, the time and space averages are assumed identical (Stull, 1988). This hypothesis is only valid for periods when the turbulent intensity is small relative to the mean wind speed (Stull, 1988), i.e. when the turbulence intensity, $I = \sigma_U / \bar{U} < 0.5$ where σ_U is the standard deviation of U. During the WASFAB campaign the turbulent intensity ranged from 0.1 to 0.35; thus Taylors Hypothesis is valid for all measurements presented here.

It is also important to check that all eddy scales contributing to the turbulent flux of particles are sampled adequately by the flux package. Very small eddies can be missed because they can occur on time scales shorter than the temporal resolution of the instrument. Likewise, large (low frequency) eddies require that the averaging time is long enough to capture their effects. Very low frequency perturbations to the flow can be caused by non-stationarity of the local flow. Non-stationarity can occur during frontal passages, rapid boundary layer growth or decay, or other short-term boundary layer disturbances such as the passage of clouds. They can be identified as periods where the time series varies in a systematic, non-turbulent, way (Massman, 2002). For these reasons, the averaging time used in flux calculations must be limited to the length of time conditions remain stationary but long enough to capture all contributing eddy scales. An averaging time of about 30 min is generally a reasonable compromise (Massman, 2002). An appropriate averaging time can be determined from a cumulative integral of the cospectrum – the so-called Ogive function. Desjardins et al. (1989) and Friehe et al. (1988) used this method to find the minimum frequency contributed by turbulent eddies. The minimum averaging time required to include all flux contributions is the inverse of this frequency. If the slope of an Ogive plot levels off at low frequencies, then all the contributing low frequency eddy scales have been captured. In each of the

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graphs in Fig. 2, the Ogive curves level off at around 0.001 Hz, a sample duration of 20 min should thus encompass all contributing eddies. This is the averaging time used in this investigation.

3.1 Effects of aerosols other than sea spray

5 To measure the production of sea spray particles using the EC method, it must be ascertained that the particles measured are indeed produced at the sea surface without contamination by non-sea spray particles. Therefore situations were selected when the wind was onshore and the air mass had resided over the ocean for at least a few days. During WASFAB, such conditions occurred between 10 and 13 October 2005. Figure 3
10 shows an example back trajectory from the NOAA Ready Hysplit model.

Clean maritime air masses might still include non sea salt particles from DMS-derived new particle formation or dust particles from the Saharan desert via long range transport. DMS-derived sulphate particle production is at its highest in the summer when stratification of the water column is greatest and the mixing depths are shallow-
15 est (Kiene, 1999), and occurs primarily over plankton blooms. DMS production can occur in October on the east US coast, however there were no plankton blooms in the area during the field campaign and chlorophyll concentrations mapped by the NASA Terra satellite were below 1 mg m^{-3} , we thus conclude there was limited biological activity to sustain local DMS production.

20 Any dust measured during this project would most likely originate in the Sahara and be transported into the atmosphere under high wind speed conditions. An examination of NASA OMI aerosol index images for the week running up to the 11 October 2005 indicates that there were no significant dust events in northern Africa. The aerosol index was consistently low for the northern part of Africa for the whole of September
25 and October 2005. Furthermore, the air mass trajectories show that the air masses sampled had been over the ocean for at least 5 days and had turned back from the Northern American continent to a westerly course over the ocean before reaching the site in Duck. Hence it is very unlikely that dust would appear in these air masses in

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quantities significant enough to influence the measurements.

Any non sea salt aerosols originating elsewhere would only contribute to the deposition flux. The net flux measured with the eddy covariance method is the sum of the upward flux of particles locally produced at the sea surface and the downward flux of particles produced elsewhere and advected into the measurement region. For particles in the size range considered here the deposition flux is small in comparison to the production flux (e.g., Slinn and Slinn, 1982; Hoppel et al., 2005), and thus unlikely to make a significant impact on the measurements.

4 Results

4.1 Summary of local meteorological and oceanic conditions

Synoptic conditions between 10 and 12 October 2005 were dominated by a weak low pressure system that tracked just offshore of the east coast of the United States, moving from south to north. As it moved north, the system deepened slightly and the wind speed increased from 3 to 12 ms⁻¹ over the three days. Figure 4 shows a summary of the local meteorological and oceanic conditions over this time period. The wind direction remained consistently between 5° and 120°, apart from a few short occasions, in particular during a short period from 03:00 p.m. till 07:00 p.m. on the 10 October; data from these periods was excluded from the analysis. The mean half-hour averaged wave heights increased from approximately 1 to 2 m over the 3 days; the mean wave period remained almost constant at approximately 10 s up to 20:00 h on 11 October, then decreased rapidly to approximately 5 s by 00:00 h 12 October; this, and an associated change in the dominant wave direction were driven by the passage of the low pressure north off shore of the measurement site. Wave periods then increased slowly to 9 seconds by the end of 12 October, with a few short intervals of longer periods, corresponding with changes in wave direction. Both the air and water temperatures decreased over the measurement period. The air temperature decreased at a rate

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faster than the water temperature. The air-sea temperature difference is negative for all but one occasion over the period of 10 to 12 October. A negative air-sea temperature difference results in an unstable lower atmosphere while a positive difference means there is warmer air over cooler water and thus a temperature inversion close to the surface. The positive air-sea temperature difference occurred at 01:00 h on the 11 October, and results from a drop in the water temperature which lasted for about 2 h; this resulted in a short period of stable conditions in the atmospheric surface layer. For the majority of the measurement period the atmosphere was either unstable or near-neutral.

4.2 Size segregated fluxes

The data were first screened for wind sector. Only periods where the instruments were all working and the local wind direction was consistently between 5° and 120° were accepted, this produced a total of 20 20-min averaged flux estimates, under a range of wind speeds from 4 to 12 ms^{-1} .

Figure 5 presents the particle fluxes associated with the accepted time intervals as a function of the mean horizontal wind speed at the 10-m level, for each group of particle sizes. Log-linear curves are fitted to the data and show the strength of the relationship – nearly all the data points in each size range falling within the 95% confidence limits. For particles around $0.5 \mu\text{m}$ radius (Fig. 5e) a correlation coefficient of $r^2=0.81$ was found for a log-linear relationship. All other size ranges have correlation coefficients greater than 0.62. The log-linear relationship between the particle fluxes and wind speed has the form

$$\text{Log}(F) = aU_{10} + b \quad (1)$$

Where F is the flux, U_{10} is the mean horizontal wind speed at 10 m and a and b are variables related to the particle size. Table 1 shows the values for a and b for each size range.

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Sea spray fluxes are also often parameterized in terms of friction velocity, u_* instead of U_{10} . An advantage of using u_* is that in principle it takes account of some of the other factors that may affect the flux, such as thermal stability and wave state (Geever et al., 2005). Relationships between the fluxes for various particle sizes and the friction velocity have been calculated and are also summarised in Table 1.

4.3 Comparison to other sea spray source functions in the literature

Figure 6 shows a comparison of the results from the WASFAB field campaign with some of the SSSF available in the literature for wind speeds of 5 m s^{-1} , 10 m s^{-1} and 12 m s^{-1} . The WASFAB spectral fluxes were calculated for each wind speed using the parameterizations presented in Table 1.

Errors in direct covariance measurements are most likely primarily due to finite time averaging and to flow distortion effects (Frederickson et al., 1997). Two types of error analysis were performed on the data. The first was to calculate the systematic errors induced by the instruments using the compound error method outlined by Blanc (1986). This method works on the principle of examining the differences between the computed variable assuming no errors and the computed variable assuming both positive and negative errors in each inputted variable. These measurement errors are associated with problems in the physical measurement of a parameter due to factors such as sensor accuracy, calibration errors, flow distortion (Frederickson et al., 1997), humidity fluctuations (Gallagher et al., 1997), particle losses down the inlet tube (Davis, 1968), etc. To calculate the systematic error associated with fluxes of particles both the error in the measured vertical wind speed and the particle concentrations needs to be decided.

The error in the vertical wind speed measured by the Solent sonic anemometer, which was used in this field work, is quoted in the manual as 1.5% for wind speeds between 0 and 60 m s^{-1} and this value has been used by Yelland et al. (1994). The error in the measured particle number concentrations for each channel of CLASP was calculated by comparing the CLASP's spectra to that of the ASASP-X. It is noted that the CLASP spectra is not consistently higher or lower than the ASASP-X which leads

to the conclusion that there is no systematic error. The overall instrument errors for the calculated flux for each CLASP channel are shown in Table 2.

The second error analysis performed was a statistical analysis. This involved calculating the standard deviation of the fluxes in each wind group and then dividing by the square root of the number of samples in the group. It is expected that the statistical errors are larger than the systematic errors because statistical error calculations are averaging together random processes and this is a limited data set. The statistical errors are overlaid on the flux results shown in Fig. 6 and are seen to be very small.

The results from this work conform well to the SSSF from the literature in both magnitude and size spectral shape, for all wind speeds. The CLASP results from the WASFAB field campaign closely follow the shape of the other functions shown in Fig. 6 in particular Monahan et al. (1986). In the two smallest size channels of CLASP the results from WASFAB indicate an increase in the number of particles with the reduction in size whereas Martensson et al. (2003), Vignati et al. (2001) and Clarke (2006) show a decrease. However, this size range is near the limit of the CLASP capability and extension to smaller sizes would be needed to evaluate the spectral behaviour of the fluxes of these particles.

The variations in the sea spray flux at different wind speeds in Figure 6 are likely due to the use of different methods, with their own inherent uncertainties, and different environmental conditions other than wind speed (e.g. wave height, wave ages, influences of the coast, stability of the atmosphere, etc.). Laboratory experiments also use different water types with varying salinity. Recent formulations have shown that u_* (Lafon) or wave height (Woolf, 2005) can provide better parameterisations for the whitecap ratio, which is a basic parameter that is often used in sea spray source function formulations (e.g. Monahan et al., 1986; Mårtensson et al., 2003).

Some of the slight discrepancies in Fig. 6 may be due to differing measurement heights and locations. Andreas (1998) applies to the flux at the surface and is based on Smith et al. (1993) which was developed from measurements at 14 m above the mean ocean surface on a sloping beach. It is noted that Smith et al. (1993) ruled out

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that these data were influenced by production in the surf zone, such surf zone production was observed by De Leeuw et al. (2000). Andreas argues that the Smith et al. SSSF underestimates the true surface production and he suggested a correction. Similarly, Smith and Harrison's measurements, for particles sized 1 to 15 μm , were at a nominal height of 10 m above the ocean surface and it is suggested that the actual SSSF right at the surface as defined by Andreas (1998) may be between 1.2 to 4 times their reported function. However, it is noted that most authors use an "effective" source height of the order of 10 m above the mean sea surface. Also different formulations are used for the particle size (cf. Lewis and Schwartz for a comprehensive discussion).

The measurements in this paper were taken at a height of 16.5 m above the ocean surface. The results were not corrected to another height because the log wind profile to be very steep due to the very low roughness length over the ocean and, thus, any correction of wind speeds for height differences will be very small. In view of the uncertainties in the flux measurements themselves these small corrections contribute little to improve the results. Andreas (2002) showed that for particles less than 2 μm in radius and for wind speeds of up to 20 m s^{-1} , the ratio between the flux at a measurement height of roughly 10 m and the flux near the ocean surface, at roughly 1 m, is very small. Therefore, there is little need to correct our measurements for height. It is worth noting that an air-sea interface source function is important for full process level knowledge but, for many processes such as the flux of particles into the mixed layer, an effective source function at the top of the constant flux layer is sufficient.

5 Conclusions

The results from the eddy covariance flux system during WASFAB show strong positive correlation between the particle fluxes and the local windspeed from 3 to 12 m s^{-1} for particles up to a micron in radius. The flux results compare favourably with a number of recent sea spray source functions from the literature across a range of wind speeds from 3 to 12 m s^{-1} . It was not, however, statistically viable to calculate an independent

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sea spray source function from this limited series of observations.

One of the main limitations with all field measurements of sea spray particles from coastal sites is that the measurements are typically representative of only one location and one point in time; frequently the statistics are also poor due to the limited quantities of data obtained. This limitation applies here – the WASFAB field campaign lasted three weeks, but good conditions for sea spray flux measurements were obtained on only 3 days. Coastal measurement sites are generally less than ideal due to effects of the surf zone – an intense local source of sea spray particles due to much increased wave breaking compared with the open ocean (De Leeuw et al., 2000). The long pier at Duck, combined with onshore wind conditions, provided a site well away from the effects of tides or the surf zone. The drag coefficient measurements made by Zappa et al. (2006) give us confidence that this site is reasonably representative of the open ocean under the conditions encountered in this study.

We have demonstrated the feasibility of making direct, size-resolved, flux measurements of sea spray aerosol via the eddy covariance technique for the first time. Work is ongoing to improve the size resolution of the instrument and to make measurements in the more challenging environment of the open ocean.

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References

Andreas, E. L.: Sea spray and the turbulent air-sea heat fluxes, J. Geophys. Res., 97(C7), 11 429–11 441, 1992.

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- Andreas, E. L.: A new Sea Spray Generation Function for Wind speeds up to 32 m s^{-1} , J. Phys. Oceanogr., 28(11), 2175–2184, 1998.
- Andreas, E. L.: A review of Sea Spray Generation Function for the Open Ocean, Atmos.-Ocean Interactions, 1, 1–46, 2002.
- 5 Angelova, M. D. and Webster, F.: Whitecap coverage from satellite measurements: A first step toward modeling the variability of oceanic whitecaps, J. Geophys. Res., 111, C03017, doi:10.1029/2005JC003158, 2006.
- Blanc, T. V.: The effects of inaccuracies in weather-ship data on bulk-derived estimates of flux, stability and sea-surface roughness, J. Atmos. Oceanic Technol., 3, 12–26, 1986.
- 10 Blanchard, D. C.: The production, distribution, and bacterial enrichment of the sea-salt aerosol, The air-sea exchange of Gases and Particles, P.S. Liss and W.G.N. Slinn, D. Reidel, Norwell, Mass, 407–454, 1983.
- Blanchard, D. C. and Woodcock, A. H.: The production, concentration, and vertical distribution of the sea-salt aerosol, Annals of the New York Academy of Sciences, 338, 330–347, 1980.
- 15 Buzorius, G., Rannk, U., Makela, J. M., Vesala, T., and Kulmala, M.: Vertical aerosols particles fluxes measured by eddy covariance technique using condensation particle counter, J. Aerosol Sci., 29(1/2), 157–171, 1998.
- Clarke, A. D., Owens, S. R., and Zhou, J.: An ultra fine sea -salt flux from breaking wave: Implications for cloud condensation nuclei in the remote marine atmosphere, J. Geophys. Res., 111, D06202, doi:10.1029/2005JD006565, 2006.
- 20 Davis, C. N.: The entry of aerosols into sampling tubes and heads, British, J. Appl. Phys., 1(2), 921–932, 1968.
- De Leeuw, G., Neele, F. P., Hill, M., Smith, M. H., and Vignati, E.: Sea spray aerosol production by waves breaking in the surf zone, J. Geophys. Res., 105(D2), 29 397–29 409, 2000.
- 25 De Leeuw, G., Moerman, M., Cohen, L., Brooks, B., Smith, M., and Vignati, E.: Aerosols, bubbles and sea spray production studies during the RED experiments, Proceedings AMS conference, Long Beach, CA, 9–13 Feb. 2003.
- De Leeuw, G., Moerman, M., Zappa, C. J., McGillis, W. R., Norris, S., and Smith, M.: Eddy Correlation Measurements of Sea Spray Aerosol Fluxes, in: Transport at the Air Sea Interface, edited by: Garbe, C. S., Handler, R. A., and Jähne, B., Springer-Verlag Berlin, Heidelberg, in press, 2007.
- 30 Desjardins, R. L., Macpherson, J. I., Schuepp, P. H., and Karanja, F.: An Evaluation of Aircraft Flux Measurements of CO_2 , Water Vapour and Sensible Heat, Boundary-Layer Meteorol.,

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- 47, 55–69, 1989.
- Fairall, C. W., Kerpert, J. D., and Holland, G. J.: The effects of Sea Spray on Surface Energy Transports over the Ocean, *Global Atmos. Ocean Syst.*, 2, 121–142, 1994a.
- Fairall, C. W., Banner, M., Morison, R., and Persson, W.: Dependence of spray flux on breaking waves, *Ocean Sciences Meeting of the American Geophysical Union*, Honolulu, HI, February, 2006.
- Frederickson, A., Davidson, K. L., and Edson, J. B.: A study of Wind Stress Determination Methods from a Ship and an Offshore Tower, *J. Atmos. Oceanic Technol.*, 14, 822–835, 1997.
- 10 Friehe, C. A., Williams, E. R., Grossman, R.: An inter-comparison of two aircraft gust probe systems., *Symposium on Lower Tropospheric Profiling: Needs and Technologies* Boulder, Colorado, American Meteorology Society, 1988.
- Gallagher, M. W., Beswick, K. M., Duyzer, J., Westrate, H., Choularton, T. W., and Hummelshoj, P.: Measurements of Aerosol fluxes to Speulder forest using a micrometeorological technique, *Atmos. Environ.*, 31(3), 359–372, 1997.
- 15 Geever, M., O'Dowd, C. D., van Ekeren, S., Flanagan, R., Nilsson, E. D., de Leeuw, G., and Rannik, U.: Submicron Sea Spray Fluxes, *Geophys. Res. Lett.*, 32, L15810, doi:10.1029/2005GL023081, 2005.
- Haywood, J. M., Ramaswamy, V., and Soden, B. J.: Tropospheric aerosol climate forcing in clear-sky satellite observations over the oceans, *Science*, 238, 1299–1303, 1999.
- 20 Hoppel, W. A., Caffrey, P. F., and Frick, G. M.: Particle deposition on water: Surface sources versus upwind sources, *J. Geophys. Res.*, 110, D10206, doi:10.1029/2004JD005148, 2005.
- IPCC, 2001: *Climate Change 2001: The Scientific Basis*. Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change, edited by: Houghton, J. T., Ding, Y., Griggs, D. J., Noguer, M., van der Linden, P. J., Dai, X., Maskell, K., and Johnson, C. A., Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 881pp.
- 25 Kaimal, J. C. and Finnigan, J. J.: *Atmospheric Boundary Layer flows: Their Structure and Measurement*, Oxford University Press, 1994.
- 30 Kiene, R. P.: Sulphur in the mix, *Nature*, 402, 363–364, 1999.
- Kristensen, L., Mann, J., Oncley, S. P., and Wyngaard, J. C.: How Close is Close Enough when Measuring Scalar Fluxes with Displaced Sensors?, *J. Atmos. Oceanic Technol.*, 14, 814–821, 1997.

EC fluxes of sea spray

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- Lewis, E. R. and Schwartz, S. E.: Sea Salt Aerosol Production – Mechanisms, Methods, Measurements, and Models, American Geophysical Union, 2004.
- Massman, W. J. and Lee, X.: Eddy covariance flux corrections and uncertainties in long term studies of carbon and energy exchanges, *Agric. Forest Meteorol.*, 113, 121–144, 2002.
- 5 Martensson, E. M., Nilsson, E. D., de Leeuw, G. Cohen, L. H., and Hansson, H. C.: Laboratory simulations and parameterization of the primary marine aerosol production, *J. Geophys. Res.-Atmos.*, 108(D9), 4297, doi:10.1029/2002JD002263 , 2003.
- Monahan, E. C., Fairall, C. W., Davidson, K. L., Boyle, P. J.: Observed inter-relationships amongst 10 m-elevation winds, oceanic whitecaps, and marine aerosols, *Quart. J. Roy. Meteorol. Soc.*, 109, 379–392, 1983.
- 10 Monahan, E. C.: Oceanic Whitecaps: Sea Surface Features Detectable via Satellite that are Indicators of the Magnitude of the Air-sea Gas Transfer Coefficient, *Proceeding of Indian Academic Science (Earth Planet Science)* 111, 315–331, 2002.
- Monahan, E. C., Spiel, D. E., and Davidson, K. L.: A model of marine aerosol generation via whitecaps and wave disruption, *Oceanic whitecaps*, edited by: Monahan, E. C. and Mac Niocaill, G., D. Reidel Publishing Company, 167–174, 1986.
- 15 Nilsson, E. D., Rannik, U., Swietlicki, E., Leck, C., Aalto, P. P., Zhou, J. and Norman, M.: Turbulent aerosol fluxes over the Arctic Ocean 2. Wind-driven sources from the sea, *J. Geophys. Res.-Atmos.*, 106(D23), 32 139–32 154, 2001.
- 20 O'Dowd, C. D., Lowe, J. A., and Smith, M. H.: Coupling of sea-salt and sulphate interactions and its impact on cloud droplet concentration predictions, *Geophys. Res. Lett.*, 26(9), 1311–1314, 1999.
- O'Dowd, C. D. and de Leeuw, G.: Marine Aerosol Production: a review of the current knowledge, *Phil. Trans. R. Soc. A*, 365, 1753–1774, doi:10.1098/rsta.2007.2043, 2007.
- 25 Reid, J. S., Jonsson, H. H., Smith, M. H., and Smirnov, A.: Evolution of the Vertical Profile and Flux of Large Sea-Salt Particles in a Coastal Zone, *J. Geophys. Res.-Atmos.*, 106(D11), 12 039–12 053, 2001.
- Schulz, M., de Leeuw, G., and Balkanski, Y.: Sea-salt aerosol source functions and emissions, in *Emissions of Atmospheric Trace Compounds*, edited by: Granier, C., Artaxo, P., and Reeves, C., Kluwer, 2004, 333–359, 2004.
- 30 Seinfeld, J. H. and Pandis, S. N.: *Atmospheric Chemistry and Physics. From Air Pollution to Climate Change*, John Wiley and Sons, Inc., 1997
- Slinn, S. A. and Slinn, W. G. N.: *Modelling of Atmospheric Particulate Deposition To Natural*

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- Waters, pp. 23–53. Atmospheric pollutants in natural waters, edited by: Eisenreich, S. J., Arbor Science, Michigan., 1981.
- Smith, M. H. and Harrison, N. M.: The Sea Spray Generation Function, J. Aerosol Sci., 29(supplement 1.), S189–S190, 1998.
- 5 Smith, M. H., Park, P. M., and Consterdine, I. E.: Marine aerosol Concentrations and estimated fluxes over the sea, J. Royal Meteorol. Soc., 119, 809–824, 1993.
- Spiel, D. E.: On the births of film drops from bubbles bursting on seawater surfaces, J. Geophys. Res., 103(C11), 24 907–24 918, doi:10.1029/98JC02233, 1998.
- 10 Stull, R. B.: An introduction to Boundary layer Meteorology, Kluwer Academic, San Diego, California, 1988.
- Vignati, E., de Leeuw, G., and Berkowicz, R.: Modeling coastal aerosol transport and effects of surf-produced aerosols on processes in the marine atmospheric boundary layer, J. Geophys. Res.-Atmos., 106(D17), 20 225–20 238, 2001.
- Woolf, D. K.: Parameterization of gas transfer velocities and sea-state-dependent wave breaking, Tellus, 57B, 87–94, 2005.
- 15 Yelland M. J., Taylor, P. K., Consterdine, I. E., and Smith, M. H.: The use of Inertial Dissipation Technique for shipboard wind stress determination, J. Atmos. Oceanic Technol., 11, 1093–1108, 1994.
- 20 Zappa, C. J., Tubiana, F. A., McGillis, W. R., Bent, J., de Leeuw, G., and Moerman, M. M.: Investigating wave processes important to air-sea fluxes using infrared techniques, Ocean Sciences Meeting of the American Geophysical Union, Honolulu, HI, February, 2006.

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Table 1. Log-Linear fit parameters for each of the different particle sizes as functions of wind speed, U_{10} , and friction velocity, u_* .

Particle radius μm	Function of U_{10}			Function of u_*	
	a	b	reg.coef	a	b
0.15	0.24	3.9	0.77	4.4	4.4
0.16	0.39	3.4	0.65	5.0	4.1
0.19	0.31	2.6	0.67	5.3	3.4
0.24	0.28	2.6	0.67	4.7	3.3
0.5	0.20	2.5	0.81	3.7	3.0
1.2	0.14	2.4	0.62	2.7	2.7

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Table 2. The Instrument and Statistical errors for each channel of the CLASP instrument as used in WASFAB 05.

CLASP channel	Instrument Errors	Statistical Errors %		
		5 ms ⁻¹	10 ms ⁻¹	12 ms ⁻¹
1	$\delta F = \pm 17\% \times F$	30	26	10
2	$\delta F = \pm 21\% \times F$	36	26	68
3	$\delta F = \pm 3\% \times F$	26	30	22
4	$\delta F = \pm 10\% \times F$	60	30	11
5	$\delta F = \pm 2\% \times F$	34	9	7
6	$\delta F = \pm 6\% \times F$	60	8	13

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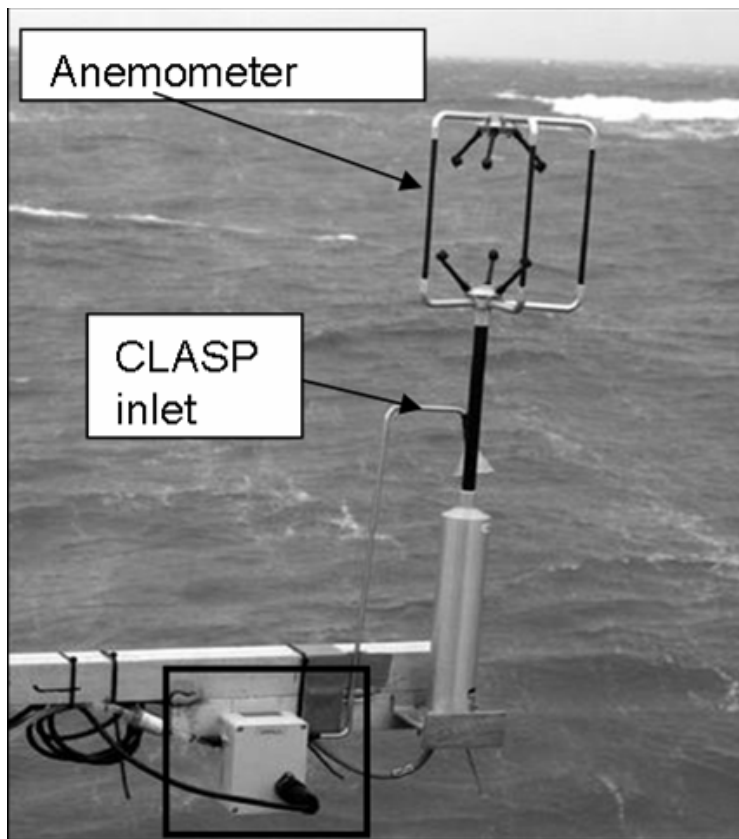


Fig. 1. Flux system consisting of the ultrasonic anemometer and the CLASP sensor (black cube) mounted at the end of a metal arm. The CLASP inlet tube can be kept short, roughly 0.5 m, because the sensor of CLASP is very small and thus does not perturb the air flow to the ultrasonic anemometer.

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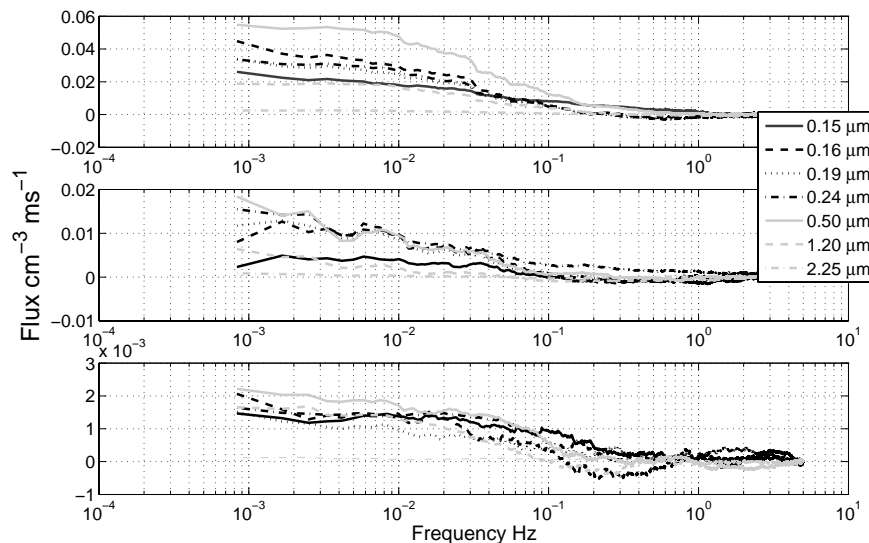


Fig. 2. Ogive plots (see text) for the cospectra between the vertical wind speed and the particle concentration for the sizes indicated in the legend for three consecutive 20 min periods from one hour period at 03:00 p.m. 24 October 2005.

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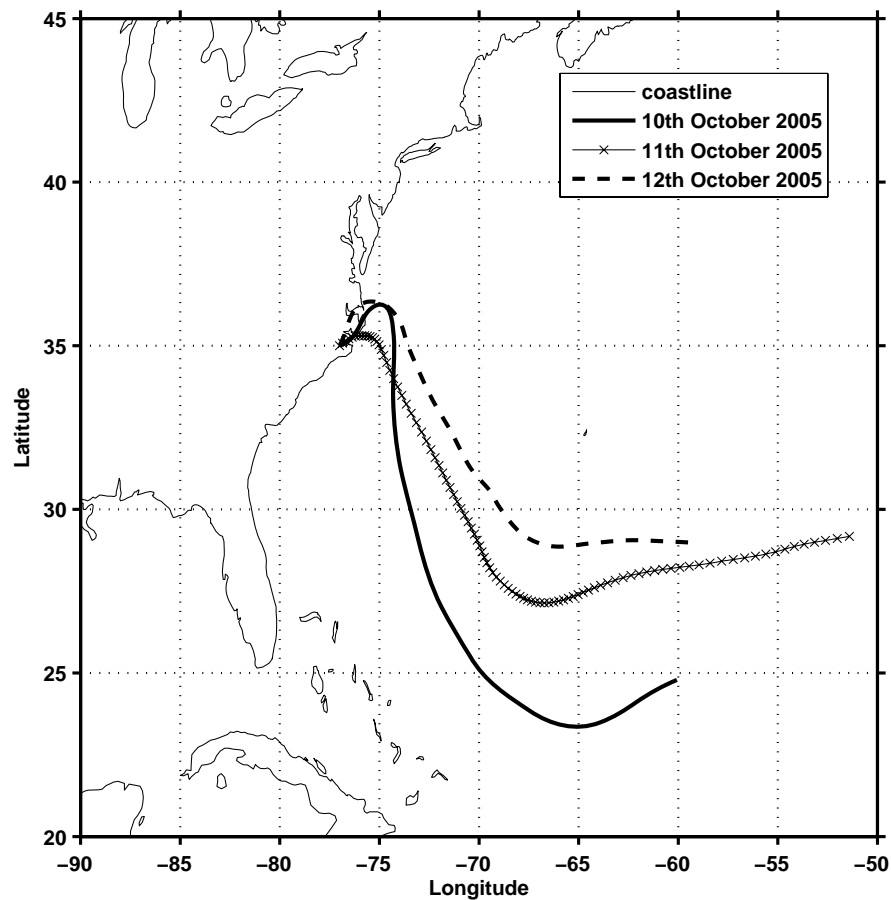


Fig. 3. Back trajectories from the NOAA Hysplit model for the 10 to 12 October 2005.

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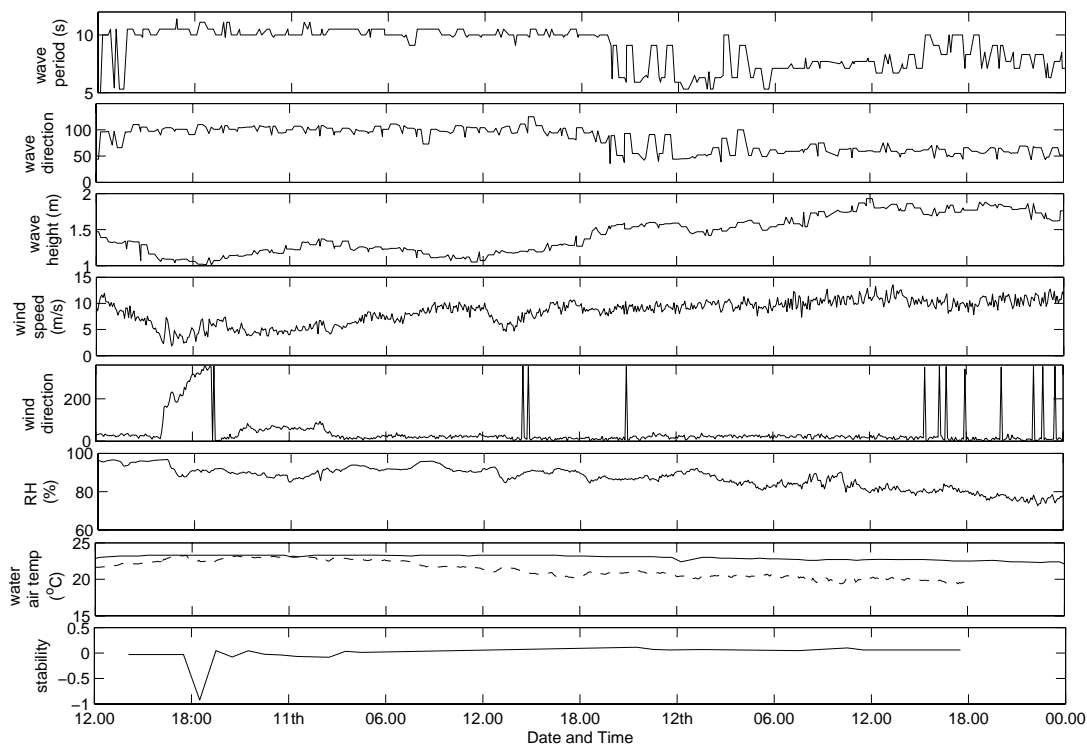


Fig. 4. Summary of the local meteorological and oceanic conditions over the onshore wind period between the 10 and the 12 October 2005 at the FRF pier in Duck, N.C. The dashed curve on the water and air temperature graph in the air temperature while the solid curve is the water temperature.

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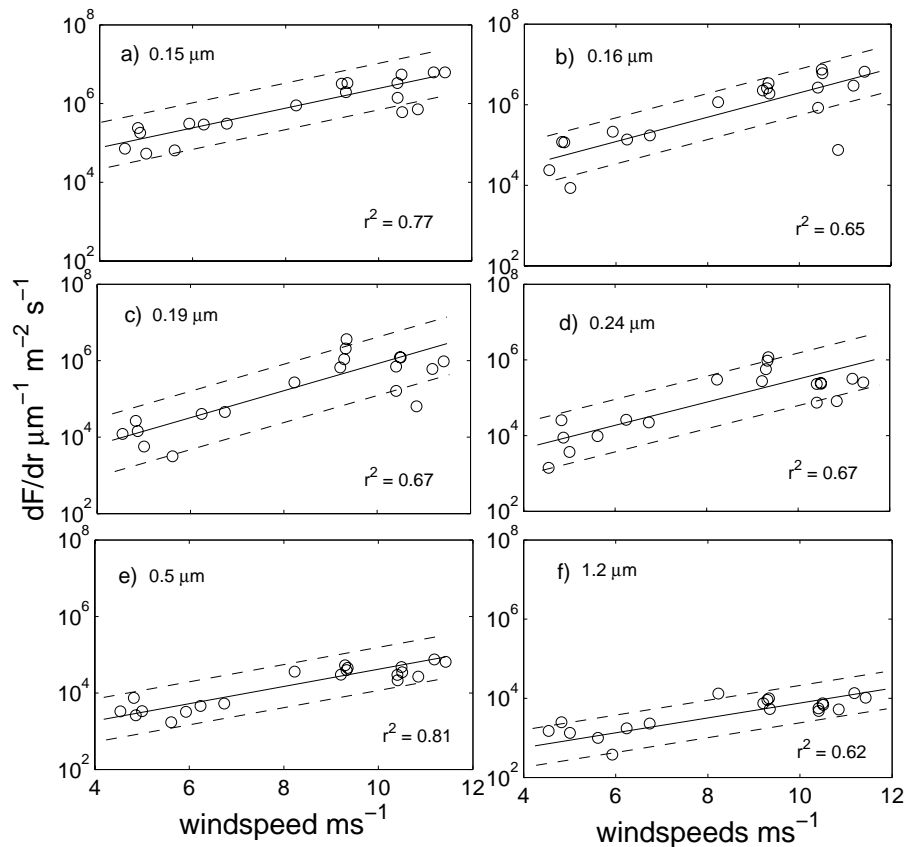


Fig. 5. Flux versus mean wind speed for mean particle radius **(a)** $0.15 \mu\text{m}$, **(b)** $0.16 \mu\text{m}$, **(c)** $0.19 \mu\text{m}$, **(d)** $0.24 \mu\text{m}$, **(e)** $0.5 \mu\text{m}$, **(f)** $1.2 \mu\text{m}$. Solid lines are log-linear fits to the data, dotted lines show the 95% confidence limits of the fit.

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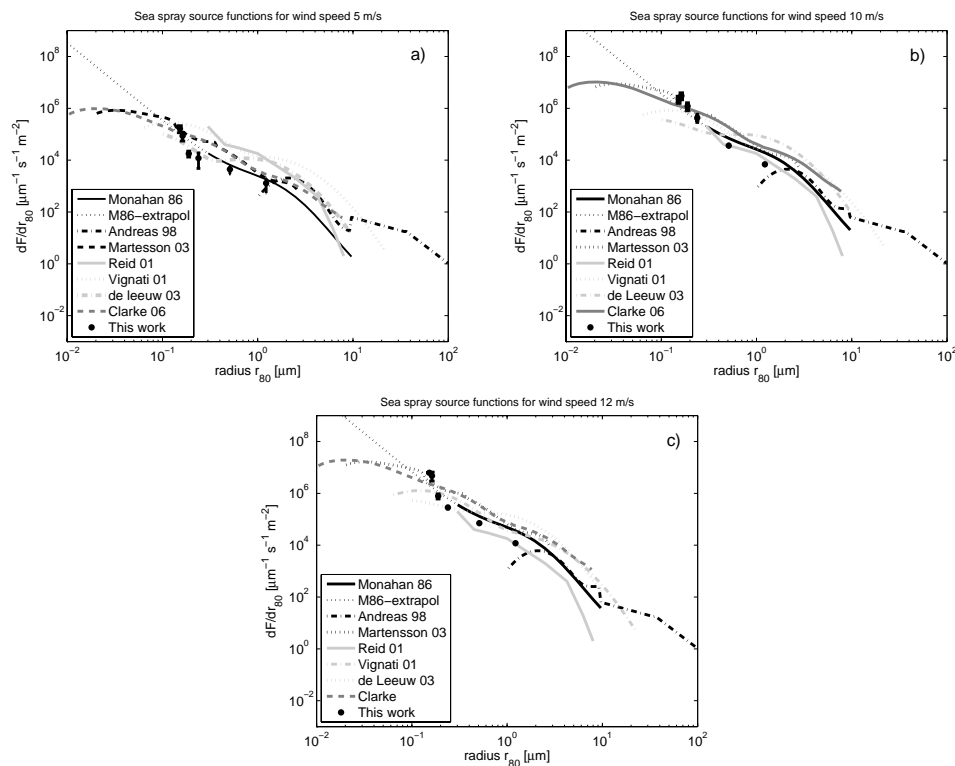


Fig. 6. Comparison of results from this work with a few sea spray source functions available in the literature for a number of different wind speeds **(a)** 5 m s^{-1} , **(b)** 10 m s^{-1} , **(c)** 12 m s^{-1} .